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SUPPLEMENTARY CEMENTITIOUS MATERIALS AND THEIR IMPACT ON SUSTAINABLE CONSTRUCTION

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ABSTRACT

As the construction sector is incessantly challenged by the growing societal demands for safer and cost-effective infrastructures, more and more environmentally-friendly products and processes have to be developed and adopted into industrial practice.

Although cementitious materials are one of the most commonly used construction materials worldwide, there are still some major concerns about their sustainability and durability. Firstly, the production of concrete is releasing large volumes of Carbon Dioxide into the atmosphere, one of the Greenhouse Gases attributable to the climate change. Secondly, even though cementitious materials are very versatile and robust they may suffer from various deteriorative processes, leading to shortened service life, and in consequence, to intrusive/expensive costs for maintenance and repair.

To meet the expectations of consumers, demanding more durable, less labour and service intensive materials at a competitive price, numerous new composite materials and technologies have been developed over the last couple of decades including blended cements with Supplementary Cementitious Materials (SCM). This paper provides a brief overview of the current situation, underlines environmental impact of these new materials and processes, and suggest some solutions for the future of construction practice. It is argued that the role of further research is critical not only in development of sustainable concrete but also in contribution to the global sustainable development.

Keywords

Sustainability of cementitious materials, Supplementary Cementitious Materials, Alkali activation, Internal curing, Copper slag, Mining wastes

PORTLAND CEMENT AND ENVIRONMENTAL IMPACT

Due to their versatility and robustness concrete and mortars based on Portland cement (PC) are the most consumed materials in buildings and infrastructure. The world cement production is growing annually and it reached 4.1 billion tonnes in 2018 [1]. This massive production of Portland cement (PC) has a major impact on the environment and the wider social and economic aspects. These, in turn result from the general availability of constituents, career opportunities, commonly accessible cement production, affordability, ability to be easily moulded into any form, mechanical and physical characteristics, as well as its long service life of concrete

structures. It is expected that the rate of the global population, especially in developing countries, will continue to increase. This major demographic shift will increase future cement demand. Figure 1 illustrates the predicted increase in cement production until 2015.

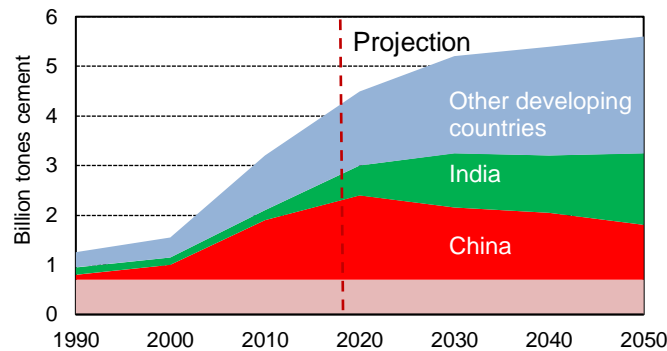


Figure 1. Analysis and forecast of cement production until 2050 [2]

Despite the wide range of motivations, a world shortage of cement production faces a number of challenges such as the abundance of resources, high temperatures during clinkering process, greenhouse gas emissions in the built environment and demolition of concrete structures.

The main concern regarding the use of cement is a vast effect on the environment. The production of Portland cement is responsible for as much as 7% of the total of global CO₂ emissions [3]. Interestingly, data compiled by the U.S. Geological Survey confirmed the relationship between GDP and environmental decay (Fig 2). The trend, known as the Environmental Kuznets Curve (EKC) reveals the link between human demand for environmental quality and their economic circumstances [4]. Societies, which are economically deficient, are more inclined to tolerate environmental degradation in order to advance their economic well-being. However, with the improvement of their economic well-being a threshold is reached where environmental issues become valued and prioritised. The Western countries are now in a position where their stage of economic development is closer to the right hand side of the graph (Fig 2.) and therefore they are more environmentally cautious.

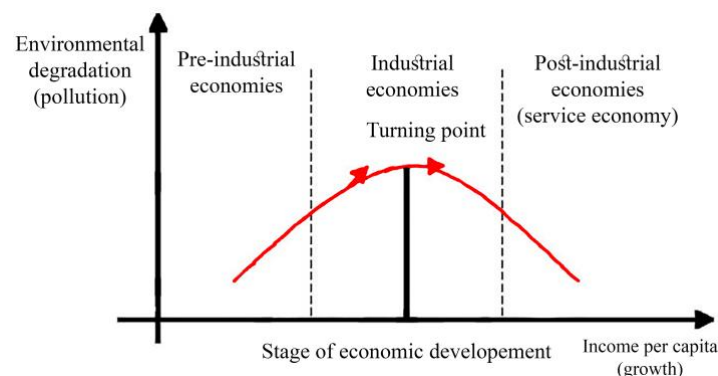


Figure 2. Environmental Kuznets Curve (EKC) – relationship between GDP and environmental decay (adapted from [4])

Hence, an enormous pressure is placed on the cement industry in developed countries to identify reliable and suitable alternative sources, technologies and energy efficiency measures, which would lead to reduction of energy consumption and CO₂ emissions.

SUPPLEMENTARY CEMENTITIOUS MATERIALS

In order to improve sustainability of Portland cement-based materials and reduce its environmental impact Supplementary Cementitious Materials (SCMs) have been widely used for a number of decades [5, 6]. SCMs can be defined as materials that, when used in conjunction with Portland cement, contribute to the properties of the hardened concrete through hydraulic or pozzolanic activity, or both. SCMs include both pozzolans and hydraulic materials and the most commonly used include fly ash, ground granulated blast furnace slag (GGBS), silica fume, and natural pozzolans, such as volcanic ash, calcined clay or shale, diatomaceous earth, rice husk ash.

Application of SCMs in concrete either in blended cements or added separately to the concrete mixer is a commonly accepted method [5]. Particularly, fly ash from coal combustion and ground granulated blast-furnace slag (GGBS) from pig iron production are regarded as a viable solution to partially substitute Portland cement [6].

The use of by-products not only contributes to cost reduction but to enhancement of concrete properties in the fresh and hardened states and to increase of service life of concrete structures [7]. For instance, workability of concrete improves notably by adding GGBS and the water demand reduces by adding 15 and 20% fly ash [8]. SCMs use up $\text{Ca}(\text{OH})_2$, product of cement hydration, to form more C-S-H and to densify the weak, $\text{Ca}(\text{OH})_2$ -laden ITZ and to reduce permeability and diffusion (secondary hydration reactions).

The use of SCMs (GGBS and fly ash) also reduces the total heat of hydration [9]. Moreover, it has been confirmed that the replacement of cement by SCMs has a positive influence on the binding capacity [7]. SCMs improve not only the resistance of concrete to chloride ion penetration (bind chloride ions in aluminates phases) [10], but also limit sulphate attack [11]. SCMs also remove excess of alkalis from pore water (binding alkalis, lower Ca/Si C-S-H) and prevent Alkali Silicate Reaction (ASR).

The SCMs reactivity depends on the chemistry of cement and SCMs, fineness, Portland composite systems, water-to-cementitious materials ratio (w/cm), and SCMs replacement level [8]. Figure 3 shows the position of main SCMs in the $\text{CaO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ ternary diagram including chemical composition.

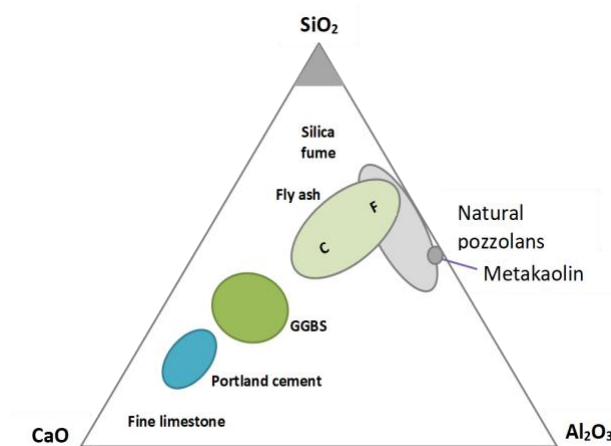


Figure 3. Chemical composition of the common SCMs [6]

The interactions between SCMs and cement hydration lead to a complex systems [6]. Many common SCMs are less reactive than PC clinker resulting in slow reactions with water. In general, total porosity increases in blended cement systems due to the slow reaction of the

SCMs, particularly at early age [7] and mechanical properties of concrete are negatively affected.

This slower initial rate of reaction is even more pronounced at high replacement levels and at lower temperatures (seasonal changes and climate effect). Some of these issues can be overcome by re-designing mixes to get early age properties, or by use of ternary mixtures—e.g. Silica fume with slag or ash; or slag mixed with high-alkali Class C fly ash.

However, very often these measures are insufficient and in order to match the pace of strength development of Portland cement, particularly during the first 14 days, alkali activation is required. In blended cements, PC clinker is essentially an activator ($\text{Ca}(\text{OH})_2$) but it is not an optimal solution. Clinker component and gypsum are balanced to give optimal rheology and strength. However, if the other material (SCM) dominates this is no longer optimal solution and some purpose-designed alkali activator should be used instead.

Since the rates of reaction decrease with decreasing temperature, the replacement levels often have to change with seasons. And just like cement, all SCMs are not the same so each type and source needs to be tested.

SUSTAINABILITY ISSUES

Fly Ash Availability and Demand

Availability of SCM as waste materials from other industries plays a critical role in the design of concrete and use of blended cements, in particular, availability of fly ash as a Coal Combustion Product (CCP). Although coal demand is expected to steadily decline in Europe, Canada, the United States and China, it is still expected to increase in India, Southeast Asia and a few other countries in Asia (Fig. 4).

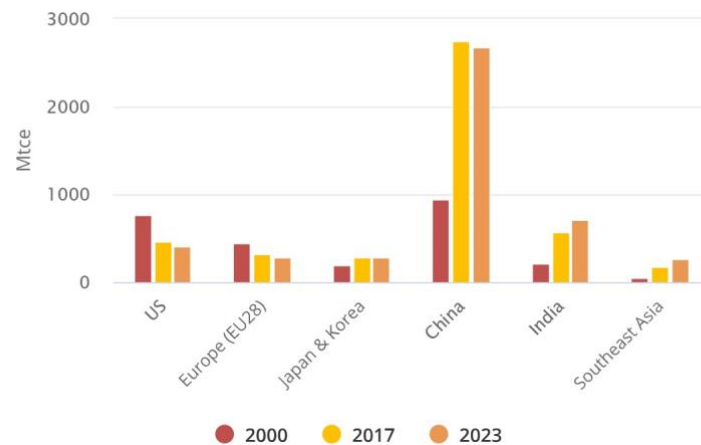


Figure 4. Coal demand in selected countries/regions in 2000, 2017 and 2023 [8]

In many advanced economies a reduction of coal-fired power generation is ongoing, in other countries a total phase out of coal power is planned over the next few decades [8]. For example in November 2015 it was announced by the UK Government that all coal fired power stations would be closed by 2025. The move away from coal in many countries is associated with an increased use of natural gas, alternative fuels such as nuclear, biomass and increased use of renewables including wind, solar, hydro and geothermal. Energy production choices and the speed of transition to alternatives depend significantly on political, economic and geographical conditions. Changing operating conditions of coal power plants leads to negative impacts on quality, consistency and availability of fly ash. It should be also noted that the availability of fly ash may significantly change with the heating season.

Carbon footprint and Alkali activation

The use of SCMs, is commonly associated with a significant reduction of carbon dioxide emissions from cement industry [9, 10]. Although a carbon footprint of concrete made with blended cements is reduced, most of SCMs used require some form of alkali activation to speed up the processes of setting and hardening. Therefore the evaluation of environmental impact should consider this issue. Indeed, many studies calculate net CO₂ savings for alkali activated binders vs Portland cement. These published values vary significantly from 9 – 97%. The realistic value is probably between 40-80% in most locations and applications [11]. However, it should be stressed that about 90% of the calculated environmental footprint is from the activator. It is particularly valid if sodium silicate is made from Na₂CO₃ produced in the Solvay process. Hence, the main current challenges are focused on reduction of CO₂ footprint of the activator and minimising the activator dose.

The comparison of environmental savings when using alkali activated ground granulated blast furnace slag in precast concrete in different applications is illustrated on Figure 5.

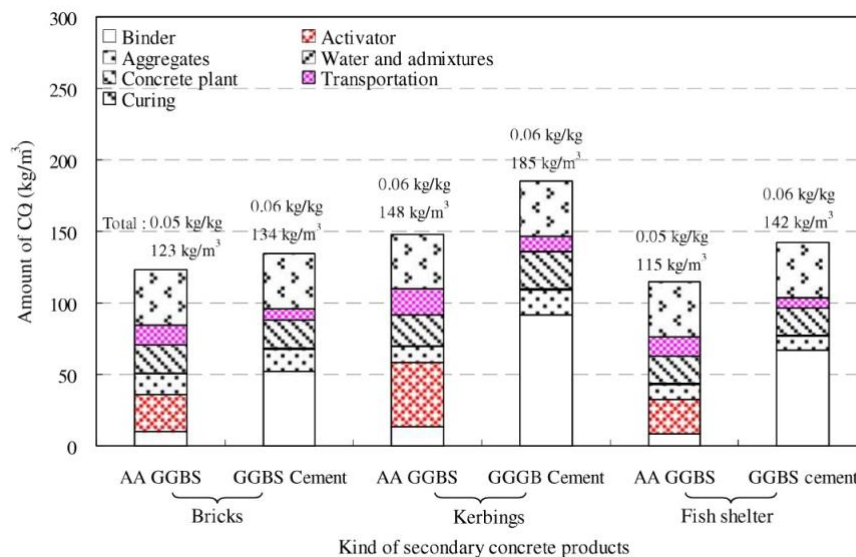


Figure 5. Assessment of CO₂ reduction of alkali-activated concrete [12]

As it can be seen the contribution of CO₂ from the cement mixes and the activator is significant. The application of the alkali activated slag cement can be worthwhile, depending on the CO₂ released during transportation and manufacture of the activator.

Selected Durability Issues

Although SCMs provide many advantages and can improve concrete performance, as mentioned earlier, they are known for their high susceptibility and sensitivity to the curing conditions.

Early age shrinkage induced by self-desiccation processes is still a major concern in such cementitious systems [13]. In an attempt to limit the autogenous shrinkage some form of internal curing may be required, for example superabsorbent polymers (SAP). SAPs are cross-linked networks of hydrophilic polymers with the ability to absorb and retain large volumes of water. Due to its high capacity to provide water-filled cavities in hardened state, SAPs may facilitate hydration processes and lead to densification of internal structure. The Figure 6 illustrates the effect of three different SAPs on autogenous shrinkage development during 180 days. The graphs refer to fibre reinforced mortars (FRM) containing three binders: CEM I 52.5N (PC) - Hanson Cement (UK), CEM II/B-V 42.5N (PC-FA) - Lafarge (UK) and CEM III/A 42.5N (PC-GGBS) - Ecocem (Ireland). SAP A, SAP C and SAP E had different water

absorption capacities (WAC) in cement paste solutions: 20 g/g, 25-30 g/g and 30 g/g respectively. The chemistry of SAP C and E were the same, but polymers had different particle sizes (E had finer particles). After the initial sharp increase in Autogenous shrinkage (AS) in all reference samples (Fig 6 a), linear changes are taking place with the diminishing rate. As expected, the influence of GGBS on AS development was very prominent.

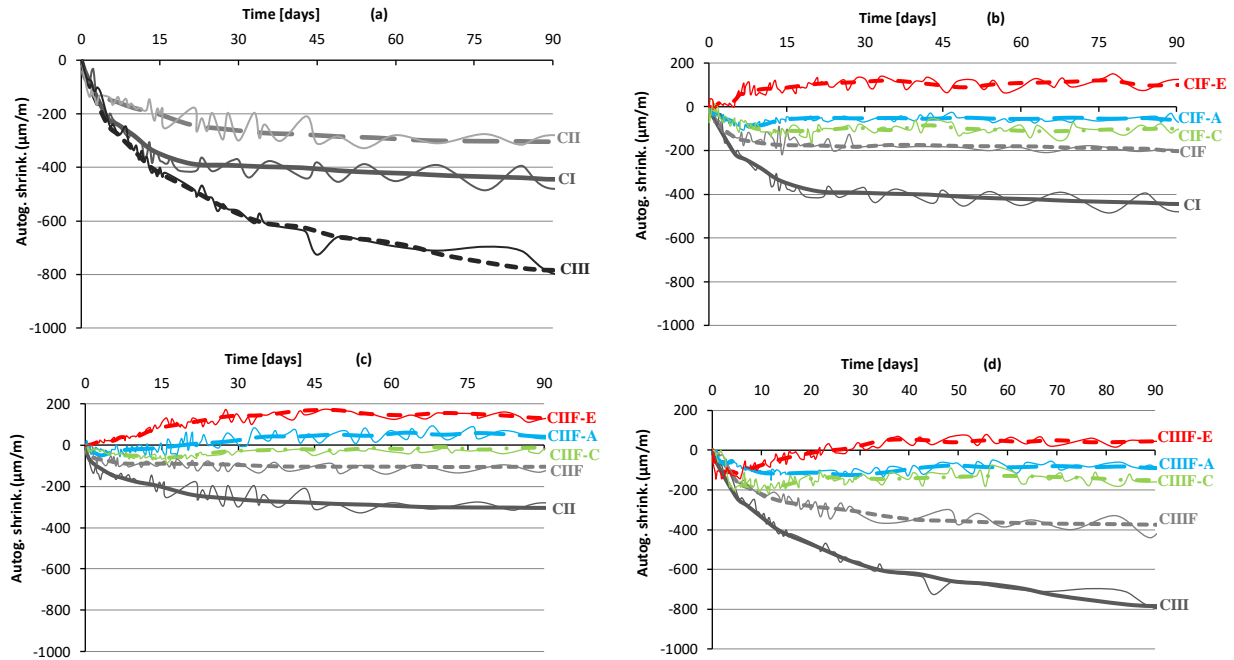


Figure 6. Reduction of AS in different blended cements by application of SAP (a) reference samples, (b) CEM I, (c) CEM II, and (d) CEM III.

Autogenous shrinkage (AS) in FRM can be completely eliminated by addition of SAPs. Although AS patterns for corresponding SAP mortars with different binders are almost identical, the extent of mitigation depends very much on the type of SAP used. SAP effectiveness depends on the particle sizes. The significant reduction of autogenous shrinkage was observed in mortars with SAP E with finer particle size. However, water absorption capacities of SAPs are of lesser importance. The effect of cement type is very limited.

The use of SAPs as internal curing agents leads to changes in kinetics and thermodynamics of cement hydration due to availability of water [14]. This in turn can contribute to the reduction of other types of deformations, for example, triggered by plastic shrinkage (PS). As shown in Fig 7 all SAPs significantly reduce PS average cracking width in all samples. As anticipated, CEM II mortars had the smallest and CEM III mortars the widest cracks widths. Increased cement fineness reduces the maximum crack width. The decrease of particle sizes of SAP can also reduce the crack widths.

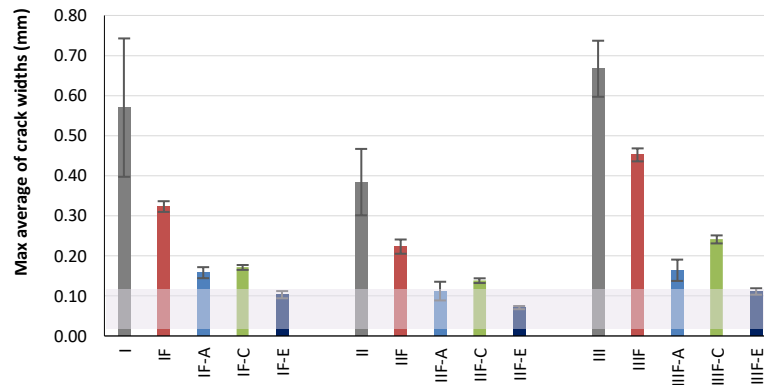


Figure 7. Maximum averages of crack widths (mm)

ALTERNATIVE SCMs

In an attempt to develop more sustainable concrete alternative SCMs, locally available waste materials, should be further investigated. As an example copper slag and mining industry wastes

Copper slag

Copper slag (CS) is an abundant material, which has been researched as a potential cement replacement [15]. Copper slag is a by-product obtained from production of copper metal, during the matte smelting and refining of copper. As copper slag contains silica and alumina, it may exhibit pozzolanic property, and hence it may be re-use in ground improvement applications as a partial replacement of cement.

The most common use of Copper slag is for abrasive applications such as blasting and grinding. This industry however cannot utilise the massive quantities in which the CS is produced. The surplus is regarded as a waste material, and commonly goes to landfill.

One of the most interesting applications of CS is the in the cement and concrete industry. CS has been investigated as a possible cement replacement due to pozzolanic activity and the ability to produce cementing properties when combined with alkali activators [16].

Copper slag is regarded as slightly less reactive than GGBS when combined with alkalis [17]. Alkali activated GGBS sets very quickly and often can cause difficulties with placement and compaction. Application of CS could be an effective way of controlling the setting times of alkali activated GGBS.

Mining wastes

Mining industry produces large amounts of waste worldwide. Waste from extractive operations involves materials that must be removed to gain access to the mineral resource, such as topsoil, overburden and waste rock, as well as tailings remaining after minerals have been largely extracted from the ore [18].

Some of these wastes are inert and hence not likely to represent a significant pollutant threat to the environment. However in many cases tailings are stored on heaps or in large ponds, where they are retained by means of dams. The collapse of dams or heaps may have serious impacts on environment and human health and safety. Examples of this are the accidents in Aberfan (Wales, 1966), Stava (Italy, 1985), Aznalcóllar (Spain, 1998), Baia Mare and Baia Borsa (Romania, 2000) and (Brazil, 2018). These impacts can have lasting environmental and socio-economic consequences and therefore wastes from the mining industries have to be properly managed.

New research on application of mining wastes in concrete [19] shows that up to 10% of the cement in concrete can be replaced by mine tailings. Tailings are the remnants of crushed ore after extraction of metals and substances. Their utilization could significantly reduce emissions of greenhouse gases from cement production. Mechanical tests of mine tailings from all over the world have documented that a large share of the world's mining waste can be used in concrete without compromising the strength of concrete. However, chemical properties of the materials must be evaluated and the environmental pollution risk should be assessed in each case.

Analyses show that while some mine tailings can be used unprocessed others have to be treated prior to application in concrete. It is also essential to determine whether heavy metals may occur in too high concentrations, and whether they could be leached out from concrete if it comes into contact with water. Pre-treatment of mining wastes may involve methods such as electrochemical cleaning and electrochemical bath. It is essential to analyse various methods of treatment and establish any potential economic and environmental gains.

Preliminary estimations reported by [19] show a significant reduction of CO₂ emissions even if mining wastes are transported over long distances.

SUMMARY

The production of concrete based on Portland cement is associated with a release of large volumes of Carbon Dioxide into the atmosphere and hence is partially responsible for the climate change. This is due to the fact that the production of concrete incorporates many intensive processes, which can cause long term environmental decay:

- Transport emissions;
- High energy demand of the rotary kiln;
- De-calcination of limestone;
- Crushing operations used to prepare solid materials.

Emissions from transport are considerable as sand, stone and water has to be transported to the processing plant or construction site. Cement clinker manufacture requires a very high temperature when it is produced, this involves burning fossil fuels. The kiln also releases large volumes of CO₂ as the crushed limestone is heated in order to drive off the CO₂ from the stone (De-calcination) which forms Calcium Oxide (CaO), one of the main constituents of cement. Crushing operations lead to even more energy consumption.

The cement industry is currently facing a major challenge to develop and adopt new measures necessary for low-carbon production and a carbon-neutral built environment. Decarbonisation of the global cement industry may require [20]

- The use of alternative raw materials and fuels to reduce the generation of CO₂ (environmental impact assessment should include all materials used, for example alkali activators)
- The use of waste heat recovery systems and clean energy, such as solar and wind power, to cut the consumption of fossil energy;
- Innovation and management optimisation in process, technology, and equipment to lower the energy consumption per unit production;
- The adoption of carbon capture technology to recycle CO₂;
- The promotion of using high-grade cement, special cement, ready-mixed concrete, and cement products to reduce cement production and cut carbon intensity.

It is most likely that the combination of the above methods and different designs for different applications may solve the problem. The “prescribed design” of concrete should be more often

replaced the “performance based design”. This is due to a variety of differences worldwide including:

- Differences in geology and availability of mineral resources;
- Differences in climate and hence different service conditions;
- Differences in economic development - development level, repair priorities and societal acceptance.

Hence the role of research and development in improvement of cement sustainability is invaluable.

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